

Phase Bifurcations of Strongly Correlated Electron Gas at the Conditions of dHvA Effect

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In a framework of catastrophe theory we investigate the equilibrium set for the system of strongly correlated electron gas at the conditions of dHvA effect and show that the discontinuities accompanied the diamagnetic phase transition (DPT) is handled by Riemann-Hugoniot catastrophe. We show that applicability of the standard condition for observation of DPT $a \geq 1$ where a is the differential magnetic susceptibility is valid only in the nearest vicinity of triple degenerate point corresponding to the center of dHvA period, but for arbitrary value of magnetic field in every period of dHvA oscillations this condition is modified in accordance with the bifurcation set of cusp catastrophe. While at the center of dHvA period the symmetric supercritical pitchfork bifurcation gives rise to the second order phase transition on temperature, the deviation of magnetic field from the value corresponding to the center of dHvA period results in the change of the phase transition order from the second to the first one both on temperature and magnetic field. In the framework of developed theory we obtain good agreement with available experimental data.

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I. INTRODUCTION

The instability of an electron gas due to strong correlations at the conditions of dHvA effect resulting in diamagnetic phase transition (DPT) into inhomogeneous diamagnetic phase (IDP) with formation of Condon domains (CDs) [1], [2] is intensively studied both theoretically and experimentally [3]-[11]. The realization of intrinsic structure of IDP is governed by the competition between long-range dipole-dipole interaction and short-range interaction related to the positive interface energy with typical magnetic length of Larmor radius. The observed IDP in Ag by splitting of NMR line [2] was identified as periodic domain structure with alternating in neighboring domains one-component magnetization. The further experiments on observation of IDP by method of μ SR spectroscopy revealed the arise of diamagnetic instability in Be, Sn, Al, Pb and In [6]-[8], giving strong support to the idea of formation of CD structure [1].

The experimental demonstration of existence of IDP in normal metals [2],[6]-[8] is very convincing and support the basic ideas of the theory of DPT developed in [1], [13], but the full understanding of the properties of IDP is still lacking. The attempts of the quantitative analysis of the data on the basis of model of plane-parallel band patterns reveal the contradictions [8], put questions on correctness of applicability of demagnetizing coefficient for adequate description of shape-dependent properties of IDP [8], [9],

[14] and raised important unanswered questions concerning the type of the diamagnetic ordering, irreversibility of DPT and morphology of the domain patterns.

There are striking similarities between IDP in normal metals and other strongly correlated systems which undergo phase transition on temperature and magnetic field with formation of complex macroscopic patterns, e. g. the type-I superconductors and thin magnetic films. The different technics, including the powder pattern and magneto-optic methods, successfully used for observation of intermediate state of type-I superconductors revealed a very rich structure [15] which in spite of all its complexity, amazingly reminds the variety of domain structures in thin film [16]. Recent experimental observation of CDs by a set of micro Hall probes at the surface of a plate-like sample of Ag [3] demonstrates the formation of complex structure which differs from the expected regular CD patterns separated by plane-parallel DWs [1] and calls for further development of the experimental techniques to study the IDP. The first experimental observation of such an exotic phenomena as diamagnetic hysteresis by complex method including direct measurements by Hall probe, standard ac method with different modulation levels, frequencies and magnetic field ramp rates [4] allows to reconstruct the magnetization reversal in Be and confirms the possibility of the first order phase transition. The giant parametric amplification of non-linear response which proves to be sensitive method for investigation of phase transitions in superconductors was observed in a single crystal of Be in quantizing magnetic field [5], where the measured output signal amazingly follows the shape of the DPT diagram and shows the asym-

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metry and shift to the upper edge of the dHvA period in ascending magnetic field justifying the possibility of the first order DPT and necessity of further development of the theory of DPT.

Motivated by recent data on observation of DPT instability [3]-[5], in the framework of catastrophe theory we investigate the equilibrium set for the system of strongly correlated electron gas at the conditions of dHvA effect. We show that in every period of dHvA oscillations the discontinuities of order parameter accompanied DPT is handled by Riemann-Hugoniot catastrophe implying the standard scenario for the transition, e.g. DPT is of the second order at the center of dHvA period, weakly first order in the nearest vicinity of this point and is of the first order at the rest part of the dHvA period both on *temperature* and *magnetic field*. Thus, similar to other magnetic systems, e. g. the spin [16] and metamagnetic ones [17], DPT can be realized in a number of different nonuniform phases depending on the shape of the sample.

At $T = 0$ K the fine, sub-band, structure of Landau level can results in DPT of the first order on *magnetic field* [18], but this effect based on treating the CD instability as an electron topological transition is negligibly small in comparing with the results obtained in the present publication.

We show that the condition for DPT occurrence $a=1$ where $a = \mu_0 \max\{\partial M/\partial B\}$ is a differential magnetic susceptibility is valid only in the nearest vicinity of the center of dHvA period, $x=0$, where x is an increment of magnetic field [13]. This condition is violated for $x \neq 0$ and is replaced by the generalized condition on a in accordance with the bifurcation set of cusp catastrophe. We discuss the conditions for realization of the DPT in the sample with taking into account the long-range dipole-dipole interaction and compare the results with available data.

II. MODEL

Equilibrium properties of strongly correlated electron gas at the conditions of dHvA effect in one-harmonic approximation can be described by Gibbs free energy $G(y; a, x) = a \cos(x + y) + \frac{1}{2}y^2$, which is a functional of two conjugated variables x and $y = -\partial_x G$ [13]. Here, the small-scale field $x = k\mu_0(H - H_a) \in [-\pi, +\pi]$ is the increment of the large-scale internal magnetic field $\mu_0 H$ and applied magnetic field $\mu_0 H_a$, y is oscillating part of reduced magnetization, $k = 2\pi F/(\mu_0 H_a)^2 = 2\pi/\Delta H$, F is the fundamental frequency of the dHvA oscillations corresponding to the extreme cross-section of Fermi-surface, and ΔH is dHvA period. Minimization of the free energy $G(y; a, x)$ with respect to y leads to well-known expression for the magnetization [13]

$$y = a \sin b \quad (1)$$

with the magnetic flux density $b = x + y$ being a function of increment of internal magnetic field x and reduced

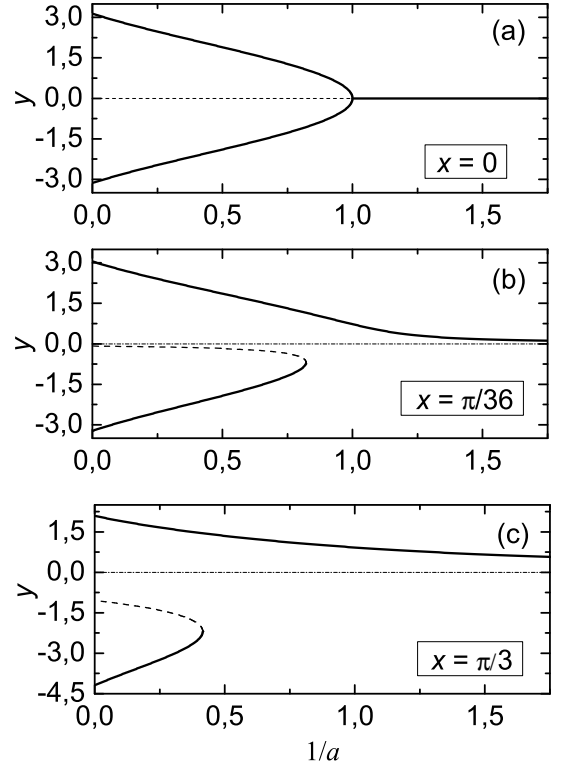


FIG. 1: Reduced magnetization y as a function of inverse differential magnetic susceptibility $1/a$ at three different values of magnetic field $x=0$ (a), $\pi/36$ (b) and $\pi/3$ (c). The solid (dash) lines correspond to the local minimum (maximum) of the free energy density $G(y; a, x)$. Symmetric supercritical pitchfork bifurcation (a) of the system undergoing the second order phase transition at the center of dHvA period $x=0$ for $a=1$ (critical point) is replaced by the imperfect bifurcation diagram (b, c) with increase of $|x| \leq \pi$ (2π is a period of dHvA oscillations in reduced units) when the system is a subject for the first order phase transition.

magnetization y . Internal magnetic field $x = x_0 + x_{ms}$ depends on applied magnetic field x_0 and stray field x_{ms} , which take into account the long-range dipole-dipole interaction dependent on the shape of a sample and has to be found by solving the Maxwell equations in magnetostatic approximation by a self-consistent procedure. As it was suggested by Shoenberg [13], the magnetic flux dependence of oscillating part of magnetization y Eq. (1) at certain conditions, when the differential susceptibility $a \geq 1$, results in arising instability of uniform phase (Shoenberg effect). At this condition both the magnetization and magnetic induction become a multi-valued function of internal magnetic field x . Therefore, depending on long-range dipole-dipole interaction at high magnetic field and low temperature the system of strongly correlated electron gas can undergo the DPT with formation of IDP which corresponds to a global minimum of the free energy of the system. Due to the governing role of the long-range dipole-dipole interactions, the morphology of the DPT depends crucially on the shape of the

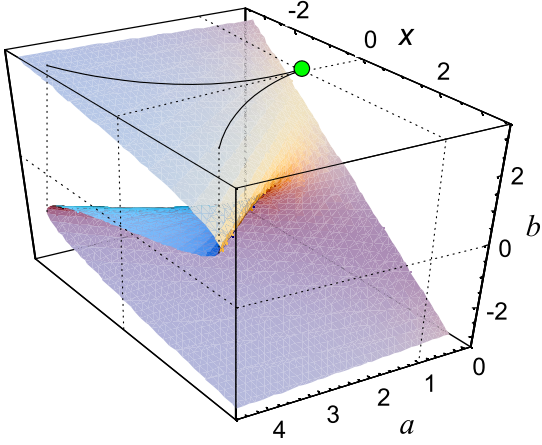


FIG. 2: (color online) Shown are the equilibrium surface and bifurcation set (solid line) of cusp catastrophe for the system of electron gas under conditions of strong dHvA effect. At crossing the bifurcation set with change of control variables a and x , the system of strongly correlated electron gas is a subject for the DPT of the first order. The only point ($a = 1, x = 0$) where the system undergoes the second-order phase transition, e. g. the critical point, is a triple degenerate point shown by solid circle.

sample which, in general, cannot be account for by the straightforward use of conception of demagnetizing coefficient [9].

The magnetization y Eq. (1) as a function of inverse reduced amplitude of the dHvA oscillations $1/a$ at three values of the increment of magnetic field $x = 0, \pi/3$ and $2\pi/3$ is plotted in Fig. 1, which illustrates the typical bifurcation behavior of the system, e. g. the existence of two stable states for magnetization at some values of the parameters $a \geq 1$ and x , and the possibility of discontinuous change of the order parameter through the disappearance of stable states when $x \neq 0$.

III. RESULTS AND DISCUSSIONS

A. Condition of Diamagnetic Instability

The discontinuities of the function $b = b(a, x)$ where parameter a controls the amount of ordering, or the value of order parameter, and parameter x breaks the Z_2 symmetry of order parameter, can be handled in the framework of catastrophe theory. The standard approach allows to obtain the canonical expression for the normal form $f(\eta; \beta) = \beta_1 - \beta_2 \eta + \eta^3$ justifying the existence of cusp catastrophe in the problem (Fig. 2). Here, the new control variables β_1 and β_2 relate to the origin ones (a, x) by means of relations $\beta_1 = -6^4 x a^{-4}$ and $\beta_2 = 6^{-3}(a-1)x a^{-4}$ correspondingly, and $\eta = 6a^{-1}b$ is the scaled phase-state variable. The direct calculations lead to the explicit form of the bifurcation set

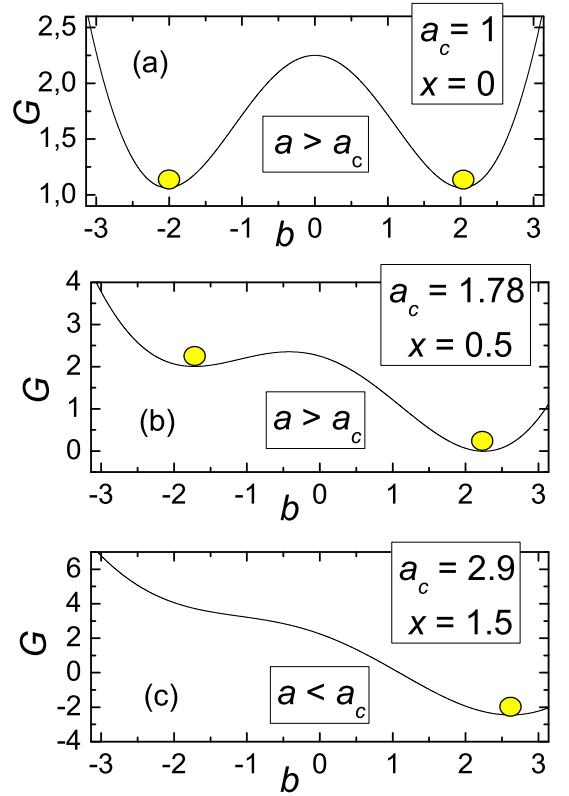


FIG. 3: (color online) Free energy density $G = G(b)$ as a function of magnetic induction for fix value $a = 2.25$ and three different value of $x = 0, 0.5$ and 1.5 is shown for one dHvA period. (a) For $x = 0$ and $a_c = 1$, the system has twofold degenerate ground state: two minima (circles) corresponding to the same value of G can give rise to the simple plane-parallel domain structure. (b,c) The increase of x followed by increase of a_c breaks the equivalency of the steady states, results in increase of the difference between the energies of two equilibrium states and possibility of realization of variety of domain structures. For $a = 2.25$ and $x = 1.5$ the only minimum implies the absolute stability of uniform diamagnetic phase.

$$a \cos(\sqrt{a^2 - 1} - |x|) = 1. \quad (2)$$

defining the bifurcation curve on (a, x) plane of control variables where the fold bifurcation occurs. Eq. (2) is a generalized condition for DPT occurrence. It can be simplified in the vicinity of triple degenerate point ($a=1, x=0$) resulting in standard cusp bifurcation set

$$8(1-a)^3 + 9x^2 = 0 \quad (3)$$

which is a semicubic parabola. It follows from Eq. (2) that the usually accepted condition for DPT occurrence $a = 1$ [13] is justified only at the center of the dHvA period, $x = 0$, but violated for $x \neq 0$ and replaced by

$$a = a_c, \quad (4)$$

where a_c is defined by Eq. (2) is a field-dependent. In particular, for $x \rightarrow 0$, we obtain

$$a_c = 1 + \frac{3^{2/3}}{2} x^{2/3} \quad (5)$$

The character of bifurcation may be seen from the surface plot $b = b(a, x)$, or the surface of stationary solutions of $b = x + a \sin b$. The results of calculations are represented in Fig. 2 which shows the equilibrium manifold of the system of strongly correlated electron gas for one period of dHvA oscillations in joint space of state and control variables (b, a, x) and the bifurcation set Eq. (2) in (a, x) -plane. At the center of dHvA period ($x = 0$) and additional condition $a \rightarrow 1$ the system exhibits the symmetric pitchfork bifurcation and undergoes the phase transition of the second order from homogeneous to inhomogeneous state at decrease of temperature T (increase of a) with formation of CD structure the type of which is governed by the long-range dipole-dipole interaction and depends on the shape of the sample. The critical point $a = 1, x = 0$, e. g. triple degenerate point, is the only point where the DPT is of the second order. But, at $x \neq 0$, a discontinuous jump of order parameter implies the possibility of existence of the first order DPT when the projection of equilibrium state on (a, x) -plane crosses the bifurcation set with change of temperature T or magnetic field x (see, also Fig. 1). A simple graphical illustration of above-mentioned effects is provided by corresponding potential function, $G(b; a, x)$, drawn as a function of magnetic induction b for $a = 2.25$ and three different value of x in Fig. 3 which shows the possibility of formation of CD structure due to existence of two minima around the center of dHvA period (Fig. 3a,b) and the absence of the condition for realization of IDP for greater values of x when two minima coalesce and the uniform ground state is not degenerate (Fig. 3c).

B. Shape Effect

The catastrophe theory allows to eliminate all the possible stable equilibria (local minima) for a given pair of control variables (a, x) with the magnetic induction b , as a phase-state variable, and establish the boundaries of CDs instability. Thus, the straightforward application of the results consists in evaluation of the range of existence of IDP, Δx_0 , defined as a part of dHvA period occupied by CDs. The boundary between different phases is governed by the bifurcation set of the cusp catastrophe. Due to the long-range character, the dipole-dipole interaction is sensitive to the shape of a sample which results in the dependence of the internal magnetic field x on experiment arrangement. The simplest way to account for the shape of the sample is the use of demagnetizing coefficient n for uniformly magnetized ellipsoid subjected to the uniform magnetic field along principal axis. In this case, the magnetization and magnetic induction are single-valued functions of applied magnetic field, x_0 , the ground state

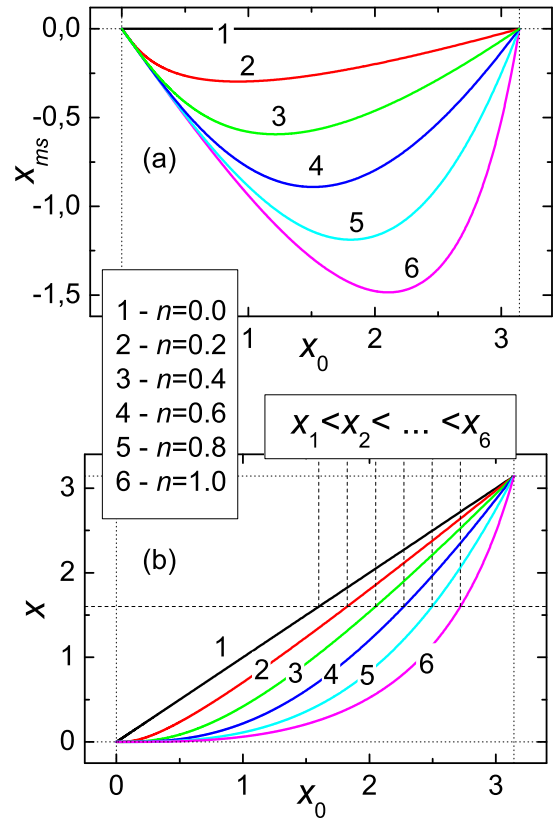


FIG. 4: (color online) (a) Magnetostatic field, x_{ms} , and (b) internal magnetic fields, x , at crossing the bifurcation set from high field side are plotted as functions of applied field, x_0 , for different values of demagnetizing coefficient, n . At given value of internal field, shown by horizontal dash line in (b), the values x_i ($i=1-6$) of applied field correspond to the increasing values of n . Both function, x_{ms} and x , are odd functions of x_0 , thus, only positive interval of x_0 is shown.

of the system is uniform and the shape effects can be calculated numerically. This procedure allows to evaluate the conditions of occurrence of the diamagnetic instability. Of course, the morphology of the domain patterns in the IDP cannot be account for by the use of the conception of demagnetizing coefficient, but has to be established by solving Maxwell equations. The results of calculation of the stray field, x_{ms} , as well as the internal field, x , as a function of applied field, x_0 , for several values of n when the uniform magnetization goes to the value defined by bifurcation set Eq. (2) are plotted in Fig. 4 which illustrates the well-known effect, e. g. the increase of n results in the growing difference between internal and applied magnetic fields due to the increase of absolute value of stray fields. In particular, considering the samples with different demagnetizing coefficients, it follows from Fig. 4(b), that at other equal conditions, the phase boundary is reached at the higher values of applied field for the sample with the larger value of n . Therefore, IDP occupies the larger part of the dHvA period for the sample with $n \rightarrow 1$ which are preferable for observation

of CD instability. In connection to this, it is worth to be mentioned that the discovery of the Condon domains was made on a plate-like sample of Ag [2] and the diamagnetic instability was observed mostly by use the sample with the values of relevant demagnetizing coefficient in the range $n \approx 0.5 - 1$ (see, [3], [8]).

The effect of "broadening" of IDP when the phase boundary is drawn in (α_1, x_0) plane is illustrated by Fig. 5 which shows the first-order phase curves for several values of demagnetizing coefficient. In the calculation we neglected the overheating-undercooling effects.

To compare the results of the theory with available experimental data on observation of CDs instability we calculate the characteristics of IDP. The formation of IDP is characterized by two measured principal parameters, the average magnetic induction splitting, δb , defined as the difference, calculated at the center of dHvA period, between the values of magnetic induction in two adjacent domains and the range of existence of CDs, δx , defined as a part of dHvA period occupied by non-uniform phase [10]. Though the occurrence of IDP and the existence of CDs itself depends on long-range dipole-dipole interaction, the average value of magnetic induction splitting, δb , depends on the differential magnetic susceptibility and with reasonable accuracy is defined in an explicit form by [13]

$$\delta b - 2a \sin \frac{\delta b}{2} = 0. \quad (6)$$

As far as concerned the range of existence of IDP δx , it is a shape-dependent quality due to dependence of internal field on dipole-dipole interaction sensitive to the sample shape, as illustrated in Fig. (4). Inasmuch as both characteristics of IDP originate from the same Eq. (1), they can be related to each other by eliminating a . It results in rather complicated functional dependence which is han-

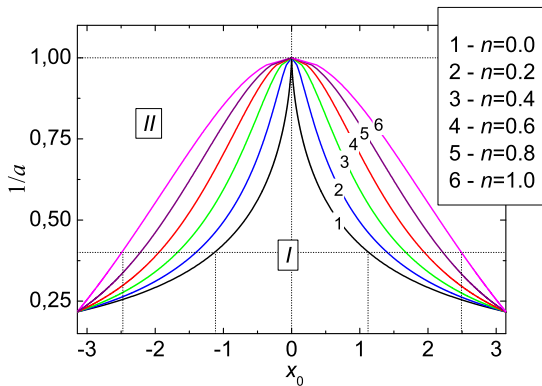


FIG. 5: (color online) The phase boundary is shown for different values of demagnetizing coefficient n in one period of dHvA oscillations. At given value of n , the inner (outer) region, designated by I (II), corresponds to non-uniform (uniform) diamagnetic phase. For fix a (horizontal dash line) the part of dHvA period occupied by IDP is wider for the sample with larger demagnetizing coefficient.

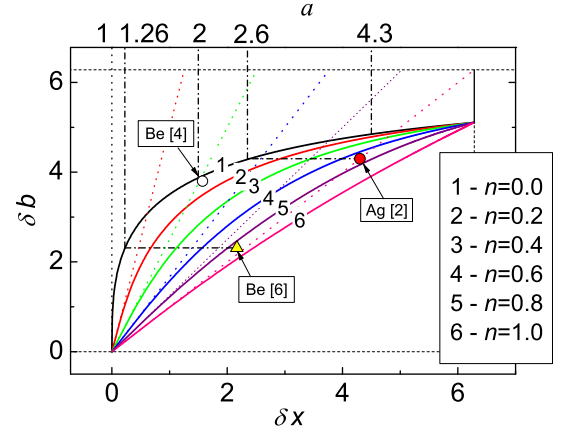


FIG. 6: (color online) Family of curves $\delta b = \delta b(\Delta x, n)$ is shown for one dHvA period. The slope of the curves at $\delta x \rightarrow 0$ is defined by inverse demagnetizing coefficient, $1/n$, in accordance with Eq. (7) justifying the Shoenberg theory of shape effects at the vicinity of critical point ($a=1, x=0$). The symbols accompanied by reference number represent the experimental data.

dled numerically, but in a limit $\delta x, \delta b \rightarrow 0$ has a simple form of linear function

$$\delta b \approx \frac{1}{n} \delta x. \quad (7)$$

From mathematical point of view Eq. (7) is fulfilled in an infinitely small vicinity of the triple degenerate point ($x=0, a=1$), justifying the applicability of Shoenberg theory of shape effect at this critical point. Accounting for the shape of the sample we calculate the family of the curves $\delta b = \delta b(\delta x, n)$ plotted in Fig. 6. The graphical representation of function $\delta b = \delta b(\delta x, n)$ which relates the characteristics of IDP to the shape of the sample is convenient to use for comparing the theoretical results with experiment. For this, we introduce also the characteristics of IDP which can be measured (or evaluated) directly in experiment, $\delta H = \delta x/k$ and $\delta B = \delta x/k$.

The formation of IDP is accompanied by irreversible behavior of the magnetization curve which can be used for experimental investigation of IDP. The experimental detection of diamagnetic hysteresis was performed on Be single crystal rod-like sample of size $8 \times 2 \times 1$ mm³ [4]. The prolate ellipsoid which approximates the sample has demagnetizing coefficient $n = 0.04$. The hysteresis was observed at ≈ 0.25 part of the dHvA period which is 2π in reduced units. It is reasonable to associate the range of existence of IDP with the range of existence of hysteresis. Thus, we obtain $\delta x \approx 1.57$. Unfortunately, the magnetic induction splitting is not reported in experiment, but it can be calculated by use of the value a due to Eq. (6). At the conditions of experiment, $T = 1.3$ K, $T_D = 2$ K (Dingle temperature) and $\mu_0 H_a = 3.6$ T, the differential magnetic susceptibility calculated in the model of slightly corrugated Fermi surface [11] is $a \approx 2$ which with the aid of Eq. (6) gives $\delta b \approx 3.8$. To verify the consistency of this

value of δb we calculate the absolute value of induction splitting $\delta B \approx 78.6$ G using the reported value of dHvA period $\Delta H = 130$ G and compare it with the experimentally detected global change of induction $\Delta B \approx 121$ G at one dHvA period. The inequality $\delta B < \Delta B$ confirms the reliability of calculated value of $\delta b \approx 3.8$. The point corresponding to the values of $\delta x \approx 1.57$ and $\delta b \approx 3.8$ is plotted in Fig. 6 and lies at the curve for $n \approx 0.04$, as it is expected.

The series of experiments on observation of CDs by μ SR spectroscopy [6]-[8] have proved the occurrence of diamagnetic instability of strongly correlated electron gas in a number of normal metals, revealed the possibility of the first-order phase transition by detecting the overheating-undercooling effects and actually served as one of the stimulation factors for development of the theory. As an example, we consider the data for Be [6] which provide the value of magnetic induction splitting and range of existence of CDs with high accuracy of $\approx \pm 1$ G in applied field of several Tesla. The μ SR measurements in [6] were performed on plate-like samples of size $1 \times 1 \times 8$ mm³. At the conditions of experiment, $T = 0.8$ K and $\mu_0 H_a = 2.75$ T, the experimental value of induction splitting $\delta B = 28.8 \pm 1.4$ G which gives $\delta b = 2.31$ with use of the reported dHvA period $\Delta H = 78.2$ G. The value of the range of existence of IDP is $\delta H \approx 27$ G or $\delta x \approx 2.2$. The plotted in Fig. 6 point, $\delta x \approx 2.2$ and $\delta b \approx 2.31$, belongs to the curve with $n \approx 0.8$ which is close to the theoretical value of $n = 0.75$ for oblate ellipsoid approximating the sample.

The famous experiment on observation of CDs instability by NMR technique [2] on the plate-like sample of Ag of size $8 \times 8 \times 0.8$ mm³ at helium temperature and applied field $\mu_0 H_a = 9$ T provides both values $\delta B = \delta H \approx 11$ which allows to calculate the reduced values $\delta b = \delta x \approx 4.32$ with use of experimental value of dHvA period $\Delta H = 15.9$ G. The corresponding point plotted in Fig. 6 lies at the curve parameterized by $n \approx 0.78$ which is close to the value $n \approx 0.84$ obtained for inscribed oblate ellipsoid approximated the sample. Thus, there is a good agreement of the theoretical results with the experimental data.

The absence of data at the vicinity of origin (Fig. 6) definitely calls for further experimental investigation of diamagnetic instability of strongly correlated electron systems.

One of the properties of the family of curves $\delta b = \delta b(\delta x, n)$ Fig. 6 consists in its universality in a sense that the curves being a graphical representation of the relationship between the measured properties of IDP (order parameter δb and range of existence of IDP δx) and characteristics of the shape of the sample (demagnetizing coefficient n) do not depend on concrete Fermi surface. Therefore, the data related to different systems exhibiting the diamagnetic instability can belong to the same curve. One can conclude that plotted in the $(\delta b, \Delta)$ plane where Δ is the range of existence of IDP in terms of internal magnetic field, the family of curves

(Fig. 6) gathers onto a single universal, or parent curve $\delta b = \delta b(\delta x, n = 0)$. This is a consequence of the universality of the bifurcation theory in description of diamagnetic instability of the electron system in a joint space (b, a, x) of phase and control variables. Another feature of the dependencies represented in Fig. 6 consists in a possibility of calculation the value of a for given experimental arrangement. As it follows from Eq. (6), a straight horizontal line in Fig. 6 crosses the family of curves in points of equal a . Mapping the points belonging to different curves onto the parent curve $\delta b = \delta b(\Delta)$ parameterized by a , with careful tabulation provides the values of a which can be compared with corresponding values calculated in different models of Fermi surface. Some examples of the mapping of the experimental points onto parent curve are shown in Fig. 6 by dash dot lines. It is follows from the analysis of the function $\delta b = \delta b(\delta x, n)$ that predicted dependency on the shape of the sample can be detectable for moderate values of reduced amplitude of dHvA oscillations $a \approx 1.3-3$. For greater values of a the high precision experiments are needed due to the small difference between the curves.

IV. CONCLUSIONS

The diamagnetic instability and stable equilibria of the system of the strongly correlated electron gas at high magnetic field and low temperature are studied by the tools of catastrophe theory. While at the center of dHvA period the symmetric supercritical pitchfork bifurcation gives rise to the second order phase transition on temperature, the deviation of magnetic field from the value corresponding to the center of dHvA period results in the change of the phase transition order from the second to the weakly first in the nearest vicinity of triple degenerate point and first at the rest part of dHvA period both on temperature and magnetic field. We calculate the generalized condition for DPT occurrence.

The results of our investigation show a reasonable agreement with the experiments. We believe also that the observation of DPT and interpretation of data can be complicated by the intrinsic structure of IDP which can realized through band patterns, bubbles and so on, but this is beyond the scope of the present publication.

We hope that our studies provide consistent framework in consideration of diamagnetic instability of strongly correlated electron gas, will help in making a quantitative systematic analysis of experimental data and stimulate the further experiments on investigation of the diamagnetic instability.

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